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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Comparison of methods for evaluation of wood smoke and estimation of UK ambient concentrations" *by* R. M. Harrison et al.

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The contribution of wood/biomass combustion to the particulate air pollution is a very important topic in air quality research, especially so for large urban areas. The contribution of Harrison et al. is therefore highly relevant and shows great promise. Comparing the different methodologies is an additional strong point of the mauscript.

The authors have used levoglucosan and water-soluble potassium as tracers of wood combustion, and compared these estimates of wood burning contribution to the aerosolized particulate matter to the ones resulting from the "Aethalometer model". The authors report a difference between the levoglucosan-based determination of

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wood-smoke and the Aethalometer model, and comment on the applicability of the Aethalometer model. I believe the application of the Aethalometer model could be enhanced with the more detailed application and further description of the details, which are now omitted from the manuscript. The authors could also compare their results with those reported for Paris (Favez 2009, Sciare 2011) and, perhaps, Grenoble (Favez 2010), all of which include systematic and detailed source apportionment efforts, where the Aethalometer model is described and compared to other source apportionment methods and measurements.

The comments below are described in the order corresponding to the data processing within the Aethalometer method, starting with the determination of source specific absorption, then proceeding to the determination of Black Carbon (BC) concentrations, apportioned to fossil fuel and wood combustion, and, finally, to the determination of the carbonaceous matter from all sources.

The authors applied the Weingartner loading compensation (Weingartner 2004) to the Aethalometer data. Several details on the methodology, which are omitted from the manuscript, would elucidate the application of the Aethalometer model. Did they use C=2,14? How did they determine the loading compensation parameter f? Was a single f used for the whole measurement period? The determination of the loading compensation parameter f is crucial. Loading compensation factors have been shown to exhibit seasonality (Virkkula 2007). If a single fixed parameter f was determined for the data whole campaign at each site (how?), this would most certainly affect the determination of the source specific contributions to BC and PM_wb. This would be most notably apparent for the measurements on the EROS site, where the campaign lasted almost one year and could, possibly explain why this site shows the unusual diurnal patterns. A short explanation on the “despiking” algorithm would also be welcome.

The authors do not report which mass absorption cross section was used to determine the BC from the absorption coefficients. Plotting the b_abs(950 nm) vs. EC would show whether the MAC is identical for all sites. The thermo-optical method for the de-

ACPD

12, C1961–C1966, 2012

termination of the OC and EC should also be reported, as this is highly relevant for the understanding of possible artefacts, leading to the systematic bias in the determination of EC (especially for wood-smoke rich samples), which consequently influences the determination of the MAC.

The source specific Angstrom exponents, used in the study, were 1,0 and 2,0 for traffic and wood combustion, respectively. Values close to these ones are used in the similar studies (all references below), however the wavelengths used in the Aethalometer model in these studies were 470 nm and 950 nm! Our experience in Nova Gorica (Slovenia) tells us that using either of the two: 370 nm or 470 nm, paired with 950 nm in the Aethalometer model gives identical results, with a notable difference: higher values of the Angstrom exponents need to be applied if using 370 nm (Močnik 2012). The explanation for this dependence of the Angstrom exponent on the wavelength is given in Moosmueller 2011. The value of the exponent used for traffic emissions is especially important and the results should be carefully examined when varying this, and other parameters. Plotting diurnal variations of source specific BC would be instructive, as well as comparing the diurnal profiles to traffic counts at major near-by roads. A sensitivity analysis would constrain the accuracy of the model. Do the rush-hour artefacts disappear at a specific value of a_ff (but see also below for additional comments on the apportionment of carbonaceous matter)?

The Aethalometer model, used to apportion the carbonaceous matter (CM), is applied in a wrong manner. The authors assume that all carbonaceous matter arises from just two sources. This is most certainly not true for urban sites. A third, non-absorbing term, should be added, using an additional parameter C_3 (see, for example, Favez 2010). This term describes the non-absorbing carbonaceous matter from sources other than fossil fuel combustion or wood burning. Because this term is omitted, all sources aside from traffic are most probably mis-apportioned to wood combustion, resulting in systematic biases. The authors conclude that the Aethalometer model reports an erroneously high contribution of wood burning, but this is not the model's fault but rather a

ACPD
12, C1961–C1966, 2012

direct consequence of the mis-application of the model. This, together with the values of the Angstrom exponent for traffic should be re-examined in great detail and results reported.

The authors should report how the coefficients C_i were determined. Compared to other campaign publications, the reported values seem low. Are coefficients C_i same for all sites? Are they same for all seasons? The determination of the three parameters C_i should be described in more detail. If multi-linear regression was used to determine them, reconstruction of CM could be reported. Seasonality should be examined at the EROS site as well. The authors report a regression between the PM_traffic and C for North Kensington. The relationship is very close, but more details should be given, and the consequences for the determination of C_i should be discussed.

It is a common misconception that the Aethalometer model apportions the primary aerosols, while in effect it does not discriminate between primary and secondary aerosols. The most absorbing portion of secondary organic aerosol (SOA) from wood emissions are humic-like substances (HULIS) and they are one of the wood-smoke components and should be reported as such. The authors' conclusion that HULIS is interference is not necessarily true, unless other sources of HULIS exist in the UK. If they do, they should be described and the Aethalometer model correspondingly modified. If there are arguments for the variable contribution of HULIS to SOA, these should be explained, and should be visible in the differences in the parameter C_2 between the sites or/and seasons. A thorough analysis of the C_i parameters is necessary and details should be reported.

A single levoglucosan to wood-smoke factor is used for all sites. This might be an oversimplification. The great range of the emission factors for levoglucosan and potassium make the use of these markers difficult with no knowledge of the type of combustion and the fuel, this is evident from the ratios between these two tracers, reported in the manuscript. The levoglucosan to wood-smoke factor also depends on both: the type of combustion and on the fuel used (not that these two can be treated completely sep-

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12, C1961–C1966, 2012

arately) – the wide range is reported by the authors and an average is used in the calculation of wood-smoke concentrations. The value is chosen arbitrarily – the distribution of the values is not necessarily normal, and the uncertainty of the analysis, depending on this value, should be discussed. An inventory of the combustion appliances, if it exists, could aid the choice of the value, but the inventories notoriously underestimate the recreational wood burning.

The most important modification to the reported use of the Aethalometer model is the third term in the sum of the source specific contributions to CM. The choice of the traffic Angstrom exponent should be examined in great detail, as it influences the Aethalometer model significantly, and the compensation explained in more detail. The levoglucosan to wood-smoke factors might be site specific and should be determined with a more thorough argumentation, and the range of possible wood-smoke concentrations using this method should be reported. A factor of about 10 has been reported for conversion of Delta C to wood-smoke (Allen, 2012) and in the presented work this agrees well with the results of the Aethalometer model, however, both of these methods could overestimate wood-smoke concentrations, as they are not independent. The Aethalometer method has been shown to give consistent results in large cities and I believe a careful examination of the raw data used in the manuscript could produce a more consistent report for the UK.

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ACPD

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